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Adjoint sensitivity of inland ozone to its precursors and meteorological and chemical influences

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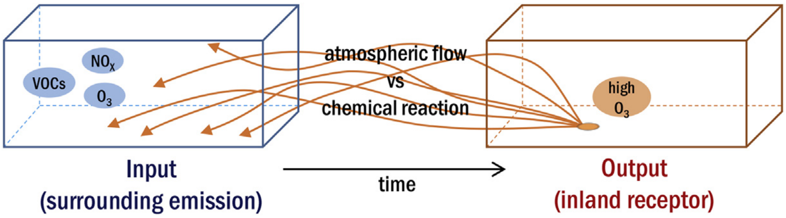
GRAPHICAL ABSTRACT



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ABSTRACT

Adjoint sensitivity analysis of NO_x, VOCs, and O_3 around the south-eastern part of the Korean Peninsula was performed to estimate their contribution to the high O_3 level in the inland city, Daegu. Weather Research and Forecasting (WRF) and Community Multiscale Air Quality (CMAQ) modeling systems were used to simulate local circulation and a high O_3 episode day. The adjoint version of the CMAQ model was applied because it is efficient in making decisions for emission control. Areas affecting day time O_3 concentration in the receptors were investigated at various times. The results show that the transport of precursors (NO_x and VOCs) was more important than that of O_3 itself. The main influence area was extended from a neighboring location to eastern coastal city Pohang, affecting mainly on the same day as the receptor time. Another sensitive area was a remote area south of Daegu, where VOCs emitted on the previous day affected O_3 concentration at the receptor time in relation with sea-breeze penetration. Sensitivities differed between the lower and upper parts of the boundary layer because of its development during the transport of O_3 precursors. After the adjoint sensitivity analysis, the influences of meteorological and chemical effects was investigated separately without the chemical reaction module; the results were 61.9% and 38.1%, respectively. This adjoint result provides valuable information for decision making regarding emission control for air quality.

Abbreviations: CMAQ, Community Multiscale Air Quality; CAPSS, Clean Air Policy Support System; DDM, Decoupled Direct Method; EPA, Environmental Protection Agency; HARC/TERC, Houston Advanced Research Center/Texas Environmental Research Consortium; IOA, Index of Agreement; KPP, Kinetic Preprocessor; LST, Local Standard Time; RMSE, Root Mean Square Error; SMOKE, Sparse Matrix Operator Kernel Emissions Model; WRF, Weather Research and Forecasting

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1. Introduction

Air pollution near the surface threatens both human health and the natural environment. In particular, predicting and understanding photochemical pollutants are difficult because chemical reactions with their precursors are complex and nonlinear. In South Korea, continuous emission control has been implemented in many industrial cities including Seoul. Since 2000, primary pollution has been showing a decreasing trend annually; however, secondary pollution has been increasing in terms of concentration and nonattainment (NIER, 2015).

Three dimensional models for air quality can be applied to understand and predict air pollution episodes and set up emission control policies effectively. These models use many input variables and parameters that determine the results to a large extent. Therefore, identifying the most important input conditions is a crucial task. In evaluating secondary pollution, the precursors of a pollutant can be quantitatively estimated by inspecting the sensitivities of model results to the emission among many input parameters.

Sensitivity analysis examines the change rate of model results with respect to input values or parameters using numerical models and can be roughly divided into two types. The first type is forward sensitivity analysis, in which the change of model output is estimated with respect to perturbation with a few input parameters at a time. It is efficient when the concentrations of all pollutants in an entire computational domain must be estimated with respect to a few emission sources at specific locations. Thus, this method is also called a source-oriented sensitivity analysis. The decoupled direct method (DDM) is well known as a representative forward sensitivity analysis, and it has been widely employed to examine the effects of changes in source emission (Cohan et al., 2005; Dunker, 1984; Dunker et al., 2002; Hakami et al., 2003). The second is adjoint (backward) sensitivity analysis, referred to as a receptor-oriented analysis. This method calculates the gradient of all the model parameter fields with the perturbation of selected receptor areas and pollutants in the reverse order of model integral. The forward sensitivity analysis cannot efficiently estimate emission sources and areas that are important for a specific receptor because the perturbed effects of all initial grid values and parameters should be separately calculated (Zhang et al., 2008). The adjoint sensitivity analysis is suitable for clarifying high concentrations of air pollutants detected at an observatory in the receptor area, whereas the forward sensitivity analysis is appropriate for studying specific events, such as a leakage of radioactive matter in a nuclear power plant (Helton, 1993; Korsakissok et al., 2013). Therefore, the receptor based sensitivity method, i.e., adjoint method is suitable for setting administrative policies about emission control, focusing on air quality around a residential area (Hakami et al., 2006).

The theoretical backgrounds of forward and adjoint sensitivity analyses were reviewed by Sandu et al. (2003). Using the kinetic preprocessor (KPP), they developed software tools for these analyses. As a companion paper of their study, Daescu et al. (2003) performed various numerical experiments to validate sensitivity results and compared consistencies between discrete and continuous adjoint analyses. Hakami et al. (2006) investigated the sensitivity of a nationwide U.S. ozone nonattainment metric with respect to precursor emissions using a continental scale chemical transport model that includes chemistry adjoint code. They revealed that NO_x and VOCs emissions contributed to ozone nonattainment by 64% and 38%, respectively, and showed the horizontal distributions of their sensitivity values. Recently, an adjoint version of the Community Multiscale Air Quality (CMAQ) model used in the U.S. Environmental Protection Agency (EPA) has been developed by Hakami et al. (2007). Comparing the adjoint method with the brute-force and DDM methods, they validated the adjoint method with numerical experiments, and suggested various applications of the adjoint sensitivity. Because of location-specific and nonlinear ozone response to NO_x emissions, Mesbah et al. (2012) proposed this highly efficient adjoint-based method to calculate the exchange rate for the cap-and-trade system. In their consecutive study (Mesbah et al., 2013), they additionally considered not only the abatement cost but also health damages to achieve socially effective emission control with the same model. Recently, they included temporal variability in the previous marginal damage of polluters using the emission behavior of power plants' based on electricity demand (Mesbah et al., 2015). Further applications regarding the monetary social and health benefits of reducing NO_x emissions are available in Pappin and Hakami (2013), and Pappin et al. (2015, 2016).

High O_3 concentrations occur not only through local emission sources but also through the transport of precursors from remote sources to the receptor area because O_3 precursors undergo photochemical reactions in due course of time. To prevent severe O_3 concentration at receptors with residential areas, the effect of the surrounding sources should be properly considered rather than only reducing emission at local sources. From this point of view, adjoint analyses are best suited to receptor-oriented applications. Further, the role of meso-scale circulation in the sensitivity analysis using the adjoint method has not been closely examined in previous studies, which mainly covered scales larger than the synoptic flow (Mallet and Sportisse, 2005; Martien et al., 2006; Pappin and Hakami, 2013; Sandu et al., 2005; Zhao et al., 2013).

In meteorology, meso-scale phenomena can be defined as those small enough to be significantly out of geostrophic wind induced by synoptic forcing but large enough that hydrostatic approximation is valid (Pielke, 2002). In the south-eastern part of South Korea, many industrial complexes are concentrated along coastal areas. The average distance between one city and another major populated city in this area is within 74 km, which is in the category of meso-scale beta, i.e. spatial and temporal dimensions of 20 km–200 km and 30 min to 6 h, respectively. Therefore, an emission source from one city clearly affects air quality in other cities through meso-scale circulation, such as land-sea breeze. Therefore, understanding air pollution in an area is difficult if only local emission sources are considered (Lee et al., 2008). In this study, we conducted the adjoint sensitivity analysis to investigate day time O_3 sensitivity with respect to NO_x, VOCs, and O_3 itself from areas around the receptor. Inland receptor areas were selected to estimate the sensitivity characteristics when local sea-breeze circulation is well developed.

2. Methods

2.1. Adjoint sensitivity

The adjoint sensitivity analysis is a technique to calculate the gradient value of the cost function defined by model output with respect to initial model parameters. Hence, it has traditionally been used for four-dimensional variational data assimilation mainly using atmospheric and oceanic numerical models. The adjoint method can be applied to air quality models to examine data assimilation as well as analyze adjoint sensitivity factors, correlating emission sources to receptor areas.

Herein, we introduce a simple expression of the sensitivity analysis rather than the entire mathematical expansion presented in Zhang et al. (2008). The numerical chemical transport model (CTM) is derived from discretized partial differential equations of atmospheric advection and diffusion with respect to space and time:

$$y^k = M(t^{k-1}, y^{k-1}, p),\ \ y^0 = y^0(t^o, p);\ \ k = 1,2,\ \ldots,\ F \tag{1}$$

where y^k is the state vector including concentration value at time t_k, M represents a solution of finite differential equation for CTM, and p is the parameter vector of model (e.g., initial and boundary values, and diffusion coefficient or emission amount). Therefore, only the state and parameters at the previous time-step determine model solution at the next step.

The sensitivity of model results at time k to the ith parameter is given by